Dynamic response of two-dimensional electron liquids in the local-density-functional theory

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The exchange-correlation potentials, $f_{xc}(q=0,\omega)$, to be used in the local-density approximation to the density-functional theory for two-dimensional homogeneous electron liquids in the normal and fully spin-polarized phases are calculated. We make use of accurate Monte Carlo energies and sum rules in the local-field correction, $G(q,\omega)$, to construct $f_{xc}(q,\omega)$ which is valid in the long-wavelength limit. Our results are qualitatively similar to the three-dimensional case.

I. INTRODUCTION

The density-functional theory proposed by Hohenberg and Kohn[1] and Kohn and Sham[2] provides an excellent tool to study the ground-state properties of condensed-matter systems. The success of local-density approximation (LDA) in time-independent problems is well known, so it is very important to develop and test its counterpart for time-dependent ones, especially in connection with response theory. Recently, Gross and Kohn[3] extended the density-functional formalism to treat the dynamic (linear) response of electron liquids within the LDA. They constructed an exchange-correlation functional that satisfied some exact conditions, including the compressibility and third-frequency moment ($\langle\omega^3\rangle$) sum rules. In their application of the formalism to three-dimensional (3D) electron liquids, using an approximate form for the $\langle\omega^3\rangle$ sum rule, correlation effects were partially neglected. Iwamoto and Gross[4] corrected the condition based on the $\langle\omega^3\rangle$ sum rule and obtained a strong frequency dependence of the exchange-correlation functional even though it was evaluated in the long-wavelength limit. A detailed review of the subject of time-dependent density-functional theory and its applications is recently given by Gross and Kohn.[5]

In the method of Gross and Kohn,[3] contact is made with the homogeneous electron gas, of which the theoretical formulation is based on the dynamic local-field correction $G(k,\omega)$. There have been several attempts to construct a dynamic $G(q,\omega)$ for the elecron gas in various approximations.[6] In relation to the time-dependent LDA, Dabrowski[7] proposed a simple parametrized form for $G(q,\omega)$ of a 3D electron liquid, which satisfies the low- and high-frequency limits exactly, and generalizes the Gross and Kohn result[3] to finite-q values. In their study of the high-frequency damping of collective excitations in fermion systems, Holas and Singwi[8] also proposed a model local-field correction $G(q,\omega)$, and formally constructed the exchange-correlation potential $f_{xc}(q,\omega)$ for a two-dimensional (2D) electron liquid. At the time, the static structure factor $S(k)$ and a parametrized form of the correlation energy $\varepsilon_c(r_s)$ from Monte Carlo (MC) simulations were not available to construct $G(q,\omega)$ explicitly.

The purpose of this article is to provide explicit expressions for the exchange-correlation potentials $f_{xc}(q=0,\omega)$ that enter the local-density approximation for 2D electron liquids both in the normal and fully spin-polarized phases. We use the results of a recent MC calculation[9] of 2D electron gas to construct the model local-field correction $G(q,\omega)$ proposed by Holas and Singwi[8] that satisfies the sum rules. Note that we are interested in the dynamic correlations in the density fluctuations of the electron liquid both in the normal (paramagnetic) and fully spin-polarized (ferromagnetic) cases. In particular, we do not investigate the dynamic response of the system due to spin fluctuations. The dynamic spin susceptibility within the framework of the time-dependent spin density-functional theory has recently been studied by Liu and Vosko.[10] In the rest of this article, we first discuss the frequency dependence of the exchange-correlation potential, $f_{xc}(q,\omega)$, within the linear-response density-functional theory. Our results for $f_{xc}(q,\omega)$ in the normal and fully spin-polarized cases are presented in Sec. III, and we conclude with a brief summary.

II. DYNAMIC RESPONSE IN THE DENSITY-FUNCTIONAL THEORY

To make this note self-contained we outline the basic steps of density-functional formalism (following closely the original work of Gross and Kohn[3]) for a 2D electron liquid, and later explicitly construct the exchange-correlation potentials required in the LDA. In the density-functional theory,[3] one considers an unperturbed homogeneous electronic system with density $n_0(\mathbf{r})$ in the ground state of the static external potential $v_0(\mathbf{r})$. For a small perturbing potential $v_1(\mathbf{r},t)$ and corresponding density $n_1(\mathbf{r},t)$, the associated Fourier components $v_1(\mathbf{r},\omega)$ and $n_1(\mathbf{r},\omega)$ are related by

$$n_1(\mathbf{r},\omega)=\int d^2r'\,\chi(|\mathbf{r}-\mathbf{r}'|;\omega)v_1(\mathbf{r}',\omega)\,, \tag{1}$$

where $\chi(|\mathbf{r}-\mathbf{r}'|;\omega)$ is the exact density-density response function. If we now assume that the density $n_0(\mathbf{r})+n_1(\mathbf{r},t)$ can be reproduced by the noninteracting system in an appropriate potential $v_0^{\rm eff}(\mathbf{r})+v_1^{\rm eff}(\mathbf{r},t)$, we can write

$$n_1(\mathbf{r},\omega) = \int d^2r'\, \chi_0(|\mathbf{r}-\mathbf{r}'|;\omega) v_1^{\rm eff}(\mathbf{r}',\omega)\,, \tag{2}$$

where χ_0 denotes dynamic response of a noninteracting system, i.e., the Lindhard function. Defining the exchange-correlation part of $v_1^{\rm eff}$ by

$$v_1^{\rm eff}(\mathbf{r},\omega) = v_1(\mathbf{r},\omega) + \int d^2r'\, \frac{n_1(\mathbf{r}',\omega)}{|\mathbf{r}-\mathbf{r}'|} + v_{1,\rm xc}(\mathbf{r},\omega)\,, \tag{3}$$

we introduce the functional relation

$$v_{1,\rm xc}(\mathbf{r},\omega) = \int d^2r'\, f_{\rm xc}(|\mathbf{r}-\mathbf{r}'|;\omega) n_1(\mathbf{r}',\omega)\,, \tag{4}$$

where the exchange-correlation potential $f_{\rm xc}$ depends on the unperturbed ground-state density $n_0(\mathbf{r})$. From the above definition of $f_{\rm xc}$, we can formally write (in Fourier space),

$$f_{\rm xc}(q,\omega) = \chi_0^{-1}(q,\omega) - \chi^{-1}(q,\omega) - v(q)\,, \tag{5}$$

where $v(q)$ is the Coulomb potential in 2D, and $\chi_0(q,\omega)$ is the dynamic susceptibility of the noninteracting system (Lindhard function). To obtain an approximation for $v_{1,\rm xc}$, the LDA is invoked, $n_0(\mathbf{r})$ and $n_1(\mathbf{r},\omega)$ are assumed to be slowly varying so that $f_{\rm xc}$ is evaluated for the local density $n_0(\mathbf{r})$, and $n_1(\mathbf{r}',\omega)$ in Eq. (4) is replaced by $n_1(\mathbf{r},\omega)$. This yields

$$v_{1,\rm xc}(\mathbf{r},\omega) \approx f_{\rm xc}(q=0,\omega;n_0(\mathbf{r})) n_1(\mathbf{r},\omega)\,. \tag{6}$$

Hence we need to find an expression for the exchange-correlation potential $f_{\rm xc}$ of the homogeneous electron liquid in the long-wavelength limit ($q \to 0$). Using the defining equation for the local-field correction $G(q,\omega)$

$$\chi(q,\omega) = \frac{\chi_0(q,\omega)}{1 + v(q)G(q,\omega)\chi_0(q,\omega)}\,, \tag{7}$$

we can make contact with the exchange-correlation potential

$$f_{\rm xc}(q,\omega) = -v(q)G(q,\omega)\,, \tag{8}$$

where the Coulomb potential reads $v(q) = 4\pi/q$ (in a.u.). The above relation holds for homogeneous systems within the linear-response theory.

Holas and Singwi[8] derived an asymptotic expression for the imaginary part of the dielectric function, $\mathrm{Im}\,\epsilon(q,\omega)$, considering a selected set of diagrams (particle-hole diagrams) in the second-order perturbation theory. They obtained the corresponding high-frequency limit for the local-field correction $G(q,\omega)$ in 2D (Ref. 8) [we use atomic units (a.u.), so that energies are in Rydbergs and lengths are in units of Bohr radius a_B],

$$\lim_{\omega\to\infty} \mathrm{Im}\, G(q,\omega) = \frac{11\pi}{32}\frac{q}{\omega}\,. \tag{9}$$

Strictly speaking the above expression is only valid at high densities, i.e., $r_s \to 0$, but it is assumed here as in the 3D case[3] to hold for a wide range of densities. Then a simple form (following the work of Gross and Kohn[3]) for $G(q,\omega)$ is proposed that interpolates between the small-ω and large-ω behaviors,

$$G(q,\omega) = G(q,\infty) + \frac{i\omega_1(q)}{\omega + i\omega_2(q)}\,, \tag{10}$$

where $\omega_1(q)$ and $\omega_2(q)$ are q-dependent functions to be determined. They are obtained by taking certain limits of $G(q,\omega)$, viz.,

$$\lim_{\omega\to\infty} [\omega\, \mathrm{Im}\, G(q,\omega)] = \omega_1(q)\,, \tag{11}$$

$$G(q,0) = G(q,\infty) + \frac{\omega_1(q)}{\omega_2(q)}\,. \tag{12}$$

Hence $\omega_1(q)$ and $\omega_2(q)$ are given by

$$\omega_1(q) = \frac{11\pi}{32} q\,, \tag{13}$$

$$\omega_2(q) = \frac{\omega_1(q)}{G(q,0) - G(q,\infty)}\,. \tag{14}$$

The high- and low-frequency limits of the local-field correction $G(q,\omega)$ at long wavelengths were calculated by Iwamoto[11] consistent with the compressibility and $\langle\omega^3\rangle$ sum rules,

$$\lim_{q\to 0} G(q,\infty) = \left[\frac{5}{6\pi}\alpha r_s + \frac{7}{8}\alpha^2 r_s^2 \varepsilon_c + \frac{19}{16}\alpha^2 r_s^3 \frac{d\varepsilon_c}{dr_s}\right] q\,, \tag{15}$$

$$\lim_{q\to 0} G(q,0) = \left[\frac{\alpha r_s}{\pi} + \frac{\alpha^2 r_s^3}{8}\frac{d\varepsilon_c}{dr_s} - \frac{\alpha^2 r_s^4}{8}\frac{d^2\varepsilon_c}{dr_s^2}\right] q\,, \tag{16}$$

where $\alpha = 2^{-1/2}$ and ε_c is the correlation energy. If we further define $f_\infty = -\lim_{q\to 0} v(q)G(q,\infty)$ and $f_0 = -\lim_{q\to 0} v(q)G(q,0)$, we can express the function $w_2(q)$ as

$$w_2(q) = \frac{(11\pi^2/8)}{f_\infty - f_0}\,. \tag{17}$$

It is clear that the exchange-correlation potential constructed above obeys all the conditions and has properties set out by Gross and Kohn[3] for the 3D case, and in particular $f_{\rm xc}$ satisfies the Kramers-Kronig relations, viz.,

$$\mathrm{Re}\, f_{\rm xc}(q,\omega) = f_\infty + \mathcal{P}\int_{-\infty}^{\infty} \frac{d\omega'}{\pi}\frac{\mathrm{Im}\, f_{\rm xc}(q,\omega')}{\omega'-\omega}\,, \tag{18}$$

$$\mathrm{Im}\, f_{\rm xc}(q,\omega) = -\mathcal{P}\int_{-\infty}^{\infty} \frac{d\omega'}{\pi}\frac{\mathrm{Re}\,[f_{\rm xc}(q,\omega')] - f_\infty}{\omega'-\omega}\,, \tag{19}$$

where $\mathcal{P}$ denotes the principal part of the integral. We have also checked the relation $f_0 < f_\infty < 0$ (for all r_s) for the MC data and observed that it is fulfilled for both the normal and the fully spin-polarized liquid phases.

III. RESULTS

We present our results of the frequency-dependent exchange-correlation potential in the long-wavelength

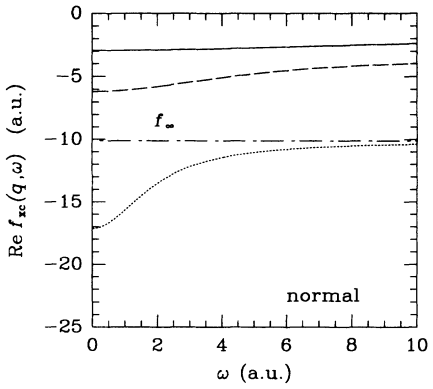

FIG. 1. Frequency dependence of the real part of the model exchange-correlation potential $f_{xc}(q = 0, \omega)$ for a 2D electron system in the normal fluid phase for $r_s = 1$ (solid line), 2 (dashed line), and 5 (dotted line).

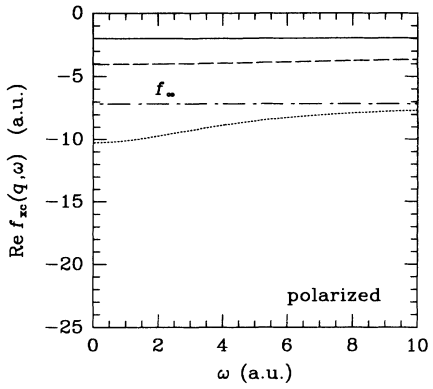

FIG. 3. Frequency dependence of the real part of the model exchange-correlation potential $f_{xc}(q = 0, \omega)$ for a 2D electron system in the spin-polarized fluid phase for $r_s = 1$ (solid line), 2 (dashed line), and 5 (dotted line).

limit, $f_{xc}(q = 0, \omega)$, in the following. We display in Fig. 1 the frequency dependence of the real part of $f_{xc}(q = 0, \omega)$ for $r_s = 1, 2$, and 5, indicated by solid, dashed, and dotted lines, respectively. Also shown is $f_\infty(r_s = 5)$ by the dotted-dashed line. We may estimate the error that would ensue in the adiabatic approximation, when $f(q = 0, \omega)$ is replaced by its value at $\omega = 0$, i.e., by f_0. For $r_s = 5$ the maximum error $\Delta = (f_0 - f_\infty)/f_0$, would be $\sim 70\%$. In practice, there exists a characteristic frequency of the system $0 < \omega^* < \infty$ for which f_{xc} is dominant and the errors of using $f_{xc}(\omega^*)$ rather than the full frequency range is considerably less than our estimate Δ. In Fig. 2 we show the frequency dependence of the imaginary part of $f_{xc}(q = 0, \omega)$ for $r_s = 1, 2$, and 5 indicated by solid, dashed, and dotted lines, respectively. The behavior of f_{xc} is qualitatively similar to the 3D case, but since $\mathrm{Im}\, f_{xc} \sim 1/\omega$ as $\omega \to \infty$, it approaches zero slower than its counterpart in 3D.

The formalism we have set out to construct the dynamic local-field correction and the exchange-correlation potential may readily be extended to calculate these quantities for the fully spin-polarized electron liquid. The correlation energy $\varepsilon_c(r_s)$ (which enters f_0 and f_∞) for the spin-polarized system obtained from a MC calculation has a parametrized form.[9] Note that we are not evaluating the spin-density response functions, hence the exchange-correlation potential is still given by Eq. (5) but the Fermi wave vector q_F that enters $\chi(q, \omega)$ is the one appropriate for the fully spin-polarized liquid. Since the Fermi wave vectors of a spin-polarized and normal liquids (in 2D) are related by $q_F^\uparrow = \sqrt{2} q_F$, the expressions for the local-field factor $G(q, \omega)$ given in Eqs. (15) and (16) are accordingly scaled. In Figs. 3 and 4 we show the real and imaginary parts of the exchange-correlation potential $f_{xc}(q = 0, \omega)$ for the fully spin-polarized electron liquid. We have plotted Figs. 3 and 4 on the same scale as Figs. 1 and 2, for easy comparison. We observe that $\mathrm{Re}\, f_{xc}$ has a somewhat stronger frequency dependence in the normal fluid phase than that in the spin-polarized case. The same is true for $\mathrm{Im}\, f_{xc}$ and the difference in the

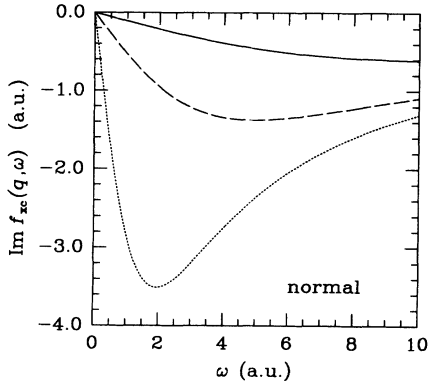

FIG. 2. Frequency dependence of the imaginary part of the model exchange-correlation potential $f_{xc}(q = 0, \omega)$ for a 2D electron system in the normal fluid phase for $r_s = 1$ (solid line), 2 (dashed line), and 5 (dotted line).

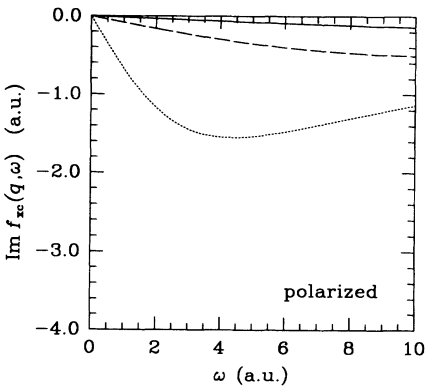

FIG. 4. Frequency dependence of the imaginary part of the model exchange-correlation potential $f_{xc}(q = 0, \omega)$ for a 2D electron system in the spin-polarized fluid phase for $r_s = 1$ (solid line), 2 (dashed line), and 5 (dotted line).

high-frequency behavior of $\mathrm{Im}f_{xc}$ for normal and spin-polarized electron liquids diminishes with increasing r_s (see Figs. 2 and 4).

IV. SUMMARY

In summary, we have presented explicit expressions for the exchange-correlation potentials $f_{xc}(q = 0, \omega)$ for 2D electron liquids in normal and spin-polarized phases as proposed by Holas and Singwi[8] and the results are qualitatively similar to the 3D case. They should be useful in future applications of the density-functional theory to 2D homogeneous electron liquids. The time-dependent density-functional theory in 3D has been successfully applied to a range of problems including the photoresponse of atoms and molecules, metallic and semiconductor surfaces, and bulk semiconductors.[12] The exchange-correlation potentials obtained here could be used in similar problems where the physical system has a 2D character, and to the extent the linear-response theory is applicable such as the calculation of atomic polarizabilities. In this work we presented a model $f_{xc}(q = 0, \omega)$ similar to the 3D case,[3,4,8] using appropriate limiting forms of $G(q, \omega)$. It would be interesting to construct a $G(q, \omega)$ with the MC structure factor $S(q)$ as input or some other scheme, such as the self-consistent field approximation of Singwi *et al.*[13] or the approach advanced by Dabrowski,[7] thereby avoiding the $q \to 0$ approximation.

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[1] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964).
[2] W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).
[3] E. K. U. Gross and W. Kohn, Phys. Rev. Lett. **55**, 2850 (1985); **57**, 923(E) (1986).
[4] N. Iwamoto and E. K. U. Gross, Phys. Rev. B **35**, 3003 (1987).
[5] E. K. U. Gross and W. Kohn, in *Advances in Quantum Chemistry* (Academic, New York, 1990), Vol. 21.
[6] S. Ichimaru, H. Iyetomi, and S. Tanaka, Phys. Rep. **149**, 91 (1987); S. Ichimaru, Rev. Mod. Phys. **54**, 1017 (1982), and references cited therein.
[7] B. Dabrowski, Phys. Rev. B **34**, 4989 (1989).
[8] A. Holas and K. S. Singwi, Phys. Rev. B **40**, 158 (1989).
[9] B. Tanatar and D. M. Ceperley, Phys. Rev. B **39**, 5005 (1989).
[10] K. L. Liu and S. H. Vosko, Can. J. Phys. **67**, 1015 (1989).
[11] N. Iwamoto, Phys. Rev. A **30**, 3289 (1984). We noticed a typographical error in this paper in Eq. (4.18a). In the second term (the term proportional to $\alpha r_s \varepsilon_c$) the numerator should read $(d^2 - 1)(4d - 1)$ instead of $4(d - 1)^2(d + 1)$.
[12] See Ref. 5 for a complete discussion of the applications of time-dependent density-functional theory.
[13] K. S. Singwi, M. P. Tosi, R. H. Land, and A. Sjölander, Phys. Rev. **176**, 589 (1968).